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Evaluation of Gamma Ray Passive Logging-Tool Calibrations for Type A Probes at the Idaho National Engineering and Environmental Laboratory Subsurface Disposal Area, OU 7-13/14

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Bechtel BWXT Idaho, LLC

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ABSTRACT

This report explores the usefulness of recalibrating logging tools used during Type A probing investigations conducted within the Subsurface Disposal Area of the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory. Limitations of the existing passive gamma-ray tool calibration method are examined in detail to assess the cause of uncertainty in quantitative results.

The passive gamma-ray tool response depends on many factors, but the three most important are (1) the amount of radionuclide source present, (2) geometry of the radionuclide source distribution (i.e., between a compact point source and a homogenous distribution), and *(3)* density of waste and soil matrix surrounding the source. Modeling exercises show these three factors have roughly equal influence on the logging-tool response. Therefore, the logging data cannot provide an unequivocal estimate for one factor unless the other factors are known. The existing calibration method solves this by making simplifying assumptions for the source geometry and soil matrix density such that the third factor can be estimated directly.

The radionuclide estimates produced by the existing calibration methodology have very limited utility because the underlying uniformity assumptions are indefensible. Inventory records, waste drum scan records, and logging data all indicate highly heterogeneous conditions within Subsurface Disposal Area waste. Furthermore, no known methods will independently measure the waste and soil matrix density. This paper considers alternative methods for gamma-ray tool calibration and data processing that acknowledge these limitations.

The proposed method would permit some nontransuranic areas to be directly identified from logging data, which may have some utility for remedial decision-making. However, this information could only be achieved at a very high monetary cost for probe installation, downhole logging, and data processing. The method is conservative by design and therefore would produce a large number of false-positive errors where nontransuranic areas are incorrectly classified as transuranic. Furthermore, no allowance is made for exotic conditions that could cause false-negative errors where transuranic waste is incorrectly classified as nontransuranic.

Finally, alternative radiation measurement technologies were investigated to determine their applicability for differentiating transuranic waste. Several large-area, long-count-time detectors were considered for deployment on the ground surface of the Subsurface Disposal Area. As a class, these tools were found to have no utility at the Subsurface Disposal Area because of the limited penetration of alpha particles, gamma rays, and neutrons through soil. New emerging radiation logging tools may significantly improve sensitivity to transuranic waste, thus allowing for much faster field operations. However, some of the new tools will suffer from the same ambiguity issues that affect existing logging tools (i.e., uncertainties about source geometry and density of the soil and waste matrix).

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ACRONYMS

DOE	U.S. Department of Energy
HPGe	high-purity germanium
INEEL	Idaho National Engineering and Environmental Laboratory
LDT	litho-density tool
PGNAA	prompt gamma neutron activation analysis
SDA	Subsurface Disposal Area
TRU	transuranic

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1. INTRODUCTION

1.1 Purpose

This report examines an alternative use for Type A probe passive gamma-ray logging data to delineate transuranic (TRU) from non-TRU waste buried in the Subsurface Disposal Area (SDA) of the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory (INEEL). Figure 1 provides a map of the INEEL showing the location of the Radioactive Waste Management Complex as well as other major areas of the INEEL. Potential approaches for calibrating logging tools and analyzing logging data for this purpose are reviewed and the limitations and uncertainties associated with these methods are discussed in this report. The review concludes that Type A probe passive gamma-ray logging data alone cannot be effectively used to define areas of TRU and non-TRU waste.

1.2 Scope

Type A passive spectral gamma-ray logging data were never intended to be used quantitatively at the SDA. Instead, their purpose was to ascertain the presence and identity of radionuclides and to map characteristic radionuclide associations within chosen study areas. The spectral gamma-ray tools were calibrated for this application to ensure that data would be consistent over time, regardless of the lapsed time between measurements or the specific tool hardware employed. The calibration functions permit tool output to be reported in units of apparent activity concentration (i.e., pCi/g or nCi/g), which can give the impression that the logging tools measure absolute radionuclide concentrations. In fact, the absolute concentrations are highly uncertain and cannot be definitely deduced from the calibrated logging data.

In this report, limitations of the existing passive gamma-ray tool-calibration method are examined in detail to assess the cause of uncertainty in quantitative results. These conclusions are used to develop an alternative calibration and data-processing method for passive gamma-ray logging data with the specific objective of differentiating TRU and non-TRU waste under SDA conditions.

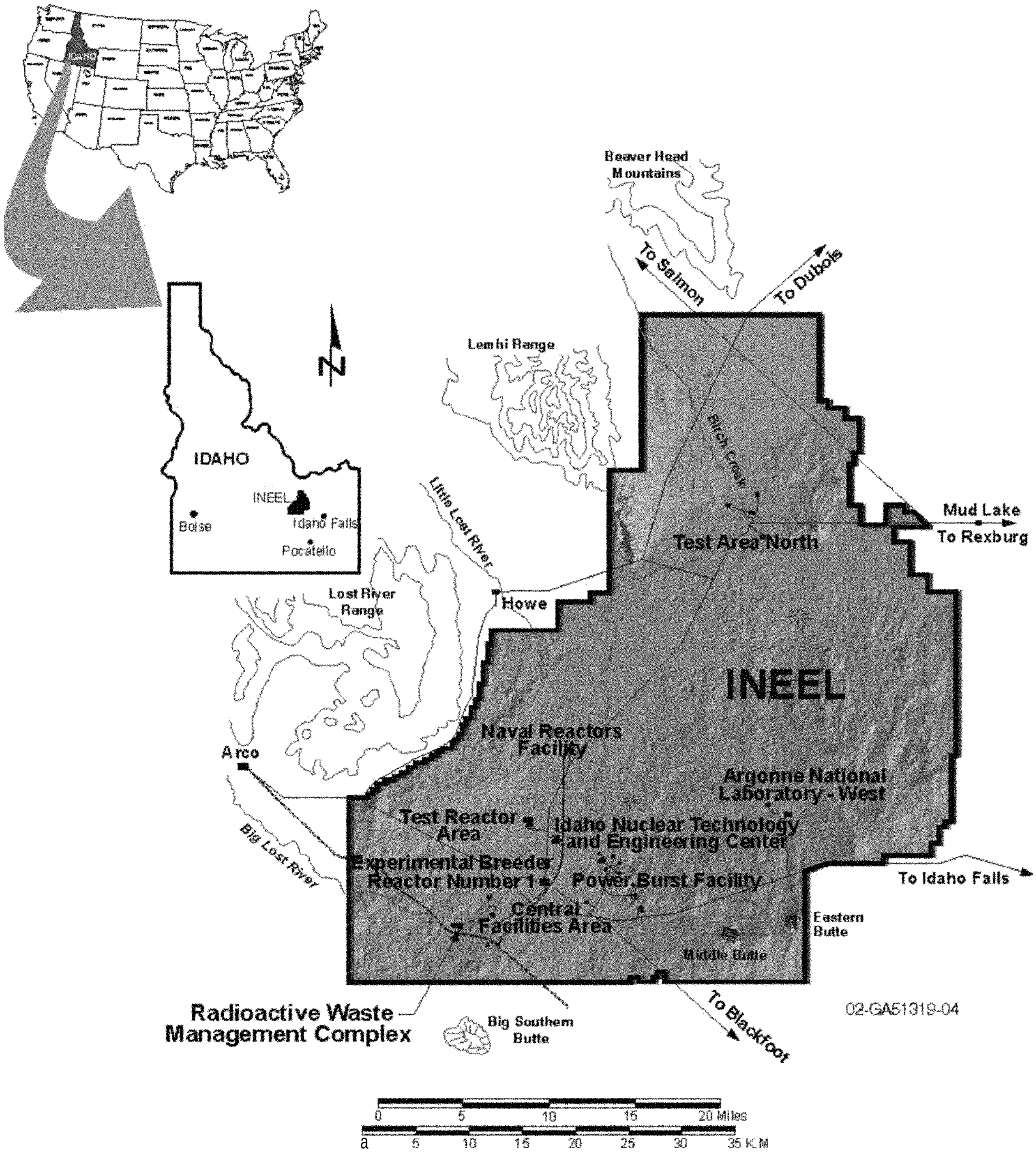

Figure 1. Map of the Idaho National Engineering and Environmental Laboratory showing the location of the Radioactive Waste Management Complex and other major Site facilities.

2. BACKGROUND

2.1 Overview of Buried-Waste Conditions at the Subsurface Disposal Area

Beginning in 1952 and continuing until 1970, radioactive and mixed waste was buried in pits and trenches at the Radioactive Waste Management Complex. Figure 2 contains a graphic presentation of the Radioactive Waste Management Complex showing the location of pits and trenches at the SDA. These types of waste were by-products of operations at the INEEL and Rocky Flats Plant"(Holdren et al. 2002) over this time period. The waste materials, packaging, and disposal methods varied substantially over time and by location within the SDA. It is difficult under these circumstances to predict typical physical and chemical conditions within the SDA waste seam. A waste retrieval test conducted from 1971 to 1972 included excavations in Pit 10 and produced some direct information concerning the physical conditions within the waste seam (Thompson 1972) as described below:

> Pit 10 was entered with the intent of retrieving Drum No. 771-3431, one of the specific drums requested for inspection by the Atomic Energy Commission. Though disposal records indicated that Pit 10 received this drum, neither Drum No. 771-3431 nor any other drum from the same load was found in Pit 10. About one-half of the drums were without identification, and about one-fourth were lidless or crushed.
>
> Random placement of the drums, which resulted from dumping, greatly complicated and slowed digging operations. Many seriously damaged drums were found, some of which were open with the inner liner ruptured. Loose material—protective mask canisters, tags, plastic material, and drum lids—was observed. The main cause of loose material and damaged drums appears to have been the practice of dumping drums. This practice also resulted in large portions of the pit volume, approximately 50%, being filled with dirt instead of waste.

These exact conditions most likely persist over much of the SDA today, including areas where Type A logging has been performed. Together, existing information on historic disposal practices and direct observations during trial waste excavations imply the likelihood of highly heterogeneous conditions in the SDA subsurface, where density and chemical composition of the waste medium may change abruptly over very short distances.

2.2 Type A Probehole Logging in the Subsurface Disposal Area

A total of 148 Type A probes were installed in or adjacent to SDA Pits 4, 5, 9, and 10 between June 1999 and July 2001. The term "Type A probe" refers to an environmentally sealed 5.5 in. outside diameter, 0.5-in. thick steel casing that can be emplaced through vadose zone soils by sonic vibratory

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a. The Rocky Flats Plant is located 26 km (16 mi) northwest of Denver. In the mid-1990s, it was renamed the Rocky Flats Environmental Technology Site. In the late 1990s, it was again renamed, to its present name, the Rocky Flats Plant Closure Project.

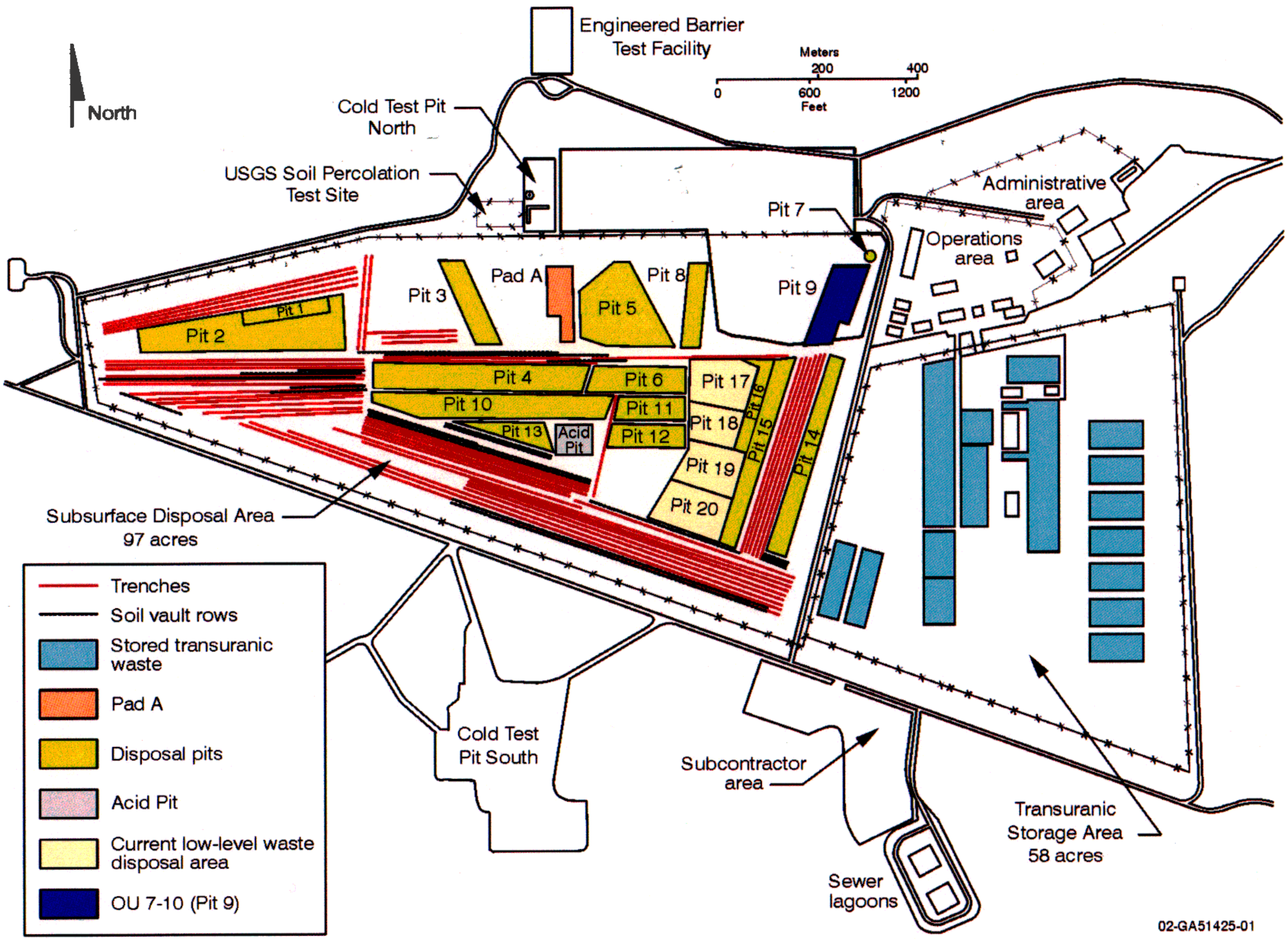

Figure 2. Graphic of the Radioactive Waste Management Complex showing pits and trenches at the Subsurface Disposal Area.

drilling methods. The Type A probes were installed specifically for deploying downhole nuclear logging tools to obtain information about the waste buried in the SDA pits. As originally conceived, the SDA logging program focused exclusively on qualitative objectives because it was uncertain whether quantitative results could be defended under the rigid guidelines for U.S. Department of Energy (DOE) cleanup projects. The original objectives included the following:

- Substantiating the location of waste shipments that have been tentatively located using waste inventory records and surface geophysics

- Identifying areas with high relative contamination levels suitable for detailed in situ studies using Type B probes

- Identifying areas with waste characteristics suitable for pilot remediation studies.

Eight exploratory logging campaigns and one investigative logging campaign were conducted, as illustrated in Figure 3. Each primary campaign targeted a specific group of waste shipments. Using five types of logging tools, over 24,000 individual logging measurements were obtained during these campaigns.

2.3 Nuclear Logging Tools

The downhole geophysical logging program relied on a standard four-tool logging suite consisting of passive spectral gamma, passive neutron, neutron-activated gamma, and soil-moisture logs. The full set of standard measurements was obtained for 135 probes. Only a partial logging suite was obtained for three probeholes in the preliminary Pit 9 campaign, and 10 probes were not logged because of shallow probe penetration. For 30 probeholes, the standard measurements were supplemented by azimuthal gamma measurements. The purpose and configuration of the logging tools are described briefly below. More detailed descriptions may be obtained from GTS Duratek reports (Meisner, Price, and Randall 2001a and 2001b; Josten and Okeson 2000).

2.3.1 Passive Spectral Gamma

The passive gamma-logging tool relies on a 35% high-purity germanium (HPGe) gamma detector. The detector, cooling system, and electronics are contained in a steel tool string having a maximum diameter of 9.21 cm (3.625 in.). The center of the detector was mounted 76.2 cm (30 in.) from the logging tool-end plug during early deployment at Pit 9, but was later changed to 10 cm (4 in.) to support measurements closer to the bottom of the probeholes. At each measurement point, the passive gamma-logging tool produces a spectral summary showing the number of gammas measured as a hnction of gamma energy. Specific radionuclides are identified by their characteristic gamma energies or those of their daughter products.

2.3.2 Neutron-Activated Spectral Gamma

Neutron-activated gamma logging employs a gamma spectrometer combined with a neutron source on the same tool string. The neutron source produces a neutron cloud in the vicinity of the probehole and the gamma spectrometer detects gammas emitted during neutron-capture reactions. The tool’s HPGe detector measures gamma flux as a hnction of energy. Certain elements (e.g., chlorine) that have a high affinity for neutron capture may be detected based on the characteristic capture gammas of that element. The neutron-activated gamma tool used at the SDA employs a 6.9-μg Cf-252 source mounted immediately above the tool end plug with the HPGe detector located 41 cm (16 in.) above the source.

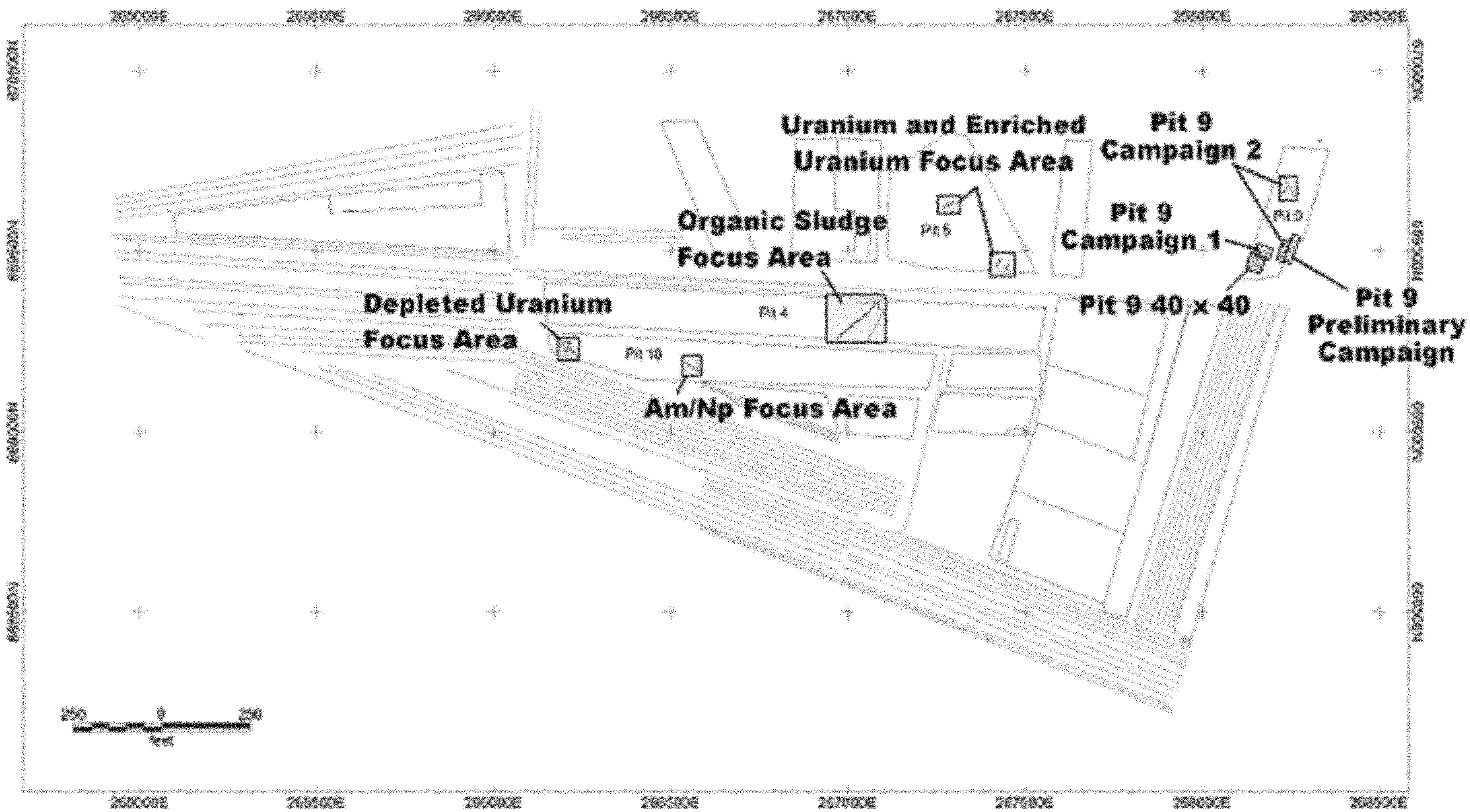

Figure 3. Exploratory and investigative logging campaigns conducted at the Subsurface Disposal Area.

Elements detected during neutron-activated gamma logging at the SDA include chlorine, iron, calcium, silicon, and hydrogen. Chlorine measurements were sought as an indicator for chlorinated volatile organic compounds. However, chlorine from other sources (e.g., plastics) cannot be distinguished from volatile organic compounds. Iron, calcium, silicon, and hydrogen provide information on the soil and waste-matrix properties.

2.3.3 Neutron-Neutron Moisture

The neutron-neutron moisture-logging tool used for SDA operations incorporates a 50-mCi americium-beryllium neutron source to irradiate the soil near the probehole and an He-3 detector to measure thermal neutron flux at a fixed distance from the source. Neutron moisture tools operate on the assumption that hydrogen atoms constitute the principal neutron moderator in the soil formation and that hydrogen is present mainly in the form of water. The moisture-logging tools used in SDA logging programs are of the close-spaced type, which means that measured count rate increases with increasing hydrogen content. The SDA logging tool employs a 3 x 13.2-cm (1 x 5.2-in.) He-3 detector with its center located 7.6 cm (3 in.) above the tool-end plug. The instrument measures count rate as a hnction of energy, but energy information is discarded during processing to produce a single value representing total counts-per-unit time.

2.3.4 Passive Neutron

Passive neutron logging employs a downhole thermal neutron detector to detect neutrons emitted by spontaneous fission and alpha-neutron reactions initiated by radionuclides within buried waste. The passive-neutron-logging tool used at the SDA employs a 5 x 12-cm (2 x 12-in.) He-3 detector. During early measurements at Pit 9, the passive neutron tool was combined on the same tool string with the passive gamma tool, but was subsequently deployed as a completely separate tool. At each measurement point, the passive neutron logging tool measures count rate as a hnction of energy, but energy information is discarded during processing to produce a single value representing total counts per unit time. The resulting bulk count rate corresponds to the steady-state neutron flux through the probehole casing at the measurement point. High neutron flux is interpreted to indicate the presence of elevated Am-241 and plutonium levels.

2.3.5 Azimuthal Spectral Gamma

The azimuthal gamma-logging tool consists of a 10% HPGe gamma spectrometer fitted with a windowed shield. After initial surveys at Pit 9, the azimuthal tool was refitted with an 18% HPGe detector and an improved shield.[b] The shield effectively blocks most gammas produced by buried waste from reaching the detector. Some gammas from buried waste pass through the shield window and are detected as a hnction of energy. Azimuthal gamma logging is performed by rotating the detector, window, and shield incrementally around the probehole and measuring a gamma spectrum at each azimuthal position. The tool permits differentiation between uniformly distributed radionuclides and radionucldes distributed as concentrated localized sources. The azimuth of the highest gamma response indicates the direction of the localized source.

In a typical azimuthal survey, measurements are initially obtained at 45-degree angular increments to identify general source direction. Additional spectra are then obtained at intermediate angles to refine the source location.

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b. The new slneld was constructed from 1.8-cm (0.7-in.) thick alloyed tungsten and provided improved gamma-ray attenuation compared with the original slneld comprising 2.1-cm (0.825-in.) tlnck mild steel.

2.4 Logging Results

Results from the SDA logging campaigns are described in several reports (Josten and Okeson 2000; Holdren et al. 2002; INEEL 2002; Josten 2002). From the beginning, logging data revealed enormous variations in subsurface conditions. While the primary observed radionuclides (i.e., plutonium, americium, neptunium, uranium, and cesium) were expected, the apparent amounts of these radionuclides changed abruptly over short distances. Depth intervals with high radionuclide indications were commonly sandwiched between layers in which the same radionuclides were undetectable. Probeholes with anomalous conditions were flanked by probeholes showing completely different anomalous conditions or no anomalies at all. The logging data revealed these heterogeneous conditions with great clarity and the subsurface representation developed from these data was well suited to meet the program's qualitative objectives. Specific information obtained from the subsurface data collected during the logging campaigns includes:

- Clearly delineated thicknesses of overburden, underburden, and waste-bearing zones.
- Specific waste types (e.g., Pu-239-bearing graphite mold waste series, Series 741 and 743 sludge, depleted uranium, and enriched uranium) were identified based on characteristics of the radionuclide mixtures and composition of the waste media. Results were consistent with waste profile information available from inventory records.
- Type A probe passive gamma-ray logging permitted Type B probes to be placed into specific waste conditions of interest.

Additionally, logging data showed that very high radiation flux zones were rare and that much of the subsurface had only low-level or undetectable radiation flux. This observation suggested the possibility that the logging data could be used to guide selective remedial actions directed at severe contamination areas only. More specifically, it was suggested that the logging data be used to differentiate TRU[c] from non-TRU waste.

These opportunities were addressed initially using the vendor-calibrated spectral gamma-ray logging data. However, attempts to use these data for quantitative purposes (e.g., to "add up" the plutonium mass in the vicinity of Probehole P9-20 [Josten 20021) quickly produced unreasonable or ambiguous results. Appropriateness of the vendor tool calibration was questioned.

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c. Transuranic is the designation used for waste that contains greater than 100 nCi/g total activity concentration due to all TRU radionuclides combined.

3. LOGGING TOOL CALIBRATION

The measurement obtained by a spectral gamma-ray logging tool in a probehole depends on many factors including:

- Gamma-emitting radionuclide distribution in the vicinity of the probehole
- Density distribution in the surrounding media
- Casing material and thickness
- Detector size and shape
- Tool housing design
- Tool signal processing electronics.

The general problem may be stated as shown in Equation (1):

$$M = F(P_1, P_2, \ldots, P_n) \tag{1}$$

where

A4	=	Count rate observed by the gamma-ray logging system
$P_1 \ldots P_n$	=	All factors that influence the count rate
F	=	Combined character of the influence that the ***P*** exert on the count rate.

This representation shows clearly that changes in measured count rate can be caused by changes in any one of the factors, ***P.*** Thus, the measurements ***A4*** have no unique interpretation in terms of any single factor ***P.***

During tool calibration, an attempt is made to isolate the influence of a single factor, ***P.*** This factor is normally some property of interest. In the present case, ***P*** is the concentration of a particular radionuclide such as Pu-239 or U-235. Influence from all field environmental factors, except ***P,*** is fixed and assumed constant. The relation is shown in Equation (2):

$$M = K \times F(P) \tag{2}$$

where

K	=	Accounts for the measurement influence of all environmental factors except ***P,*** the radionuclide concentration of interest.

The value of K and the hnction F are determined from the calibration process. Then, by applying the calibration hnction, the field measurements ***A4*** (in counts/second) may be interpreted directly in terms of radionuclide concentration estimates. The calibration hnction determined by this method is valid for any field measurements made under the conditions where K and F are valid. These conditions are called the calibration conditions or the calibration assumptions and are preferably chosen to represent the average expected conditions at the field site. When field environmental conditions vary from calibration assumptions, the true radionuclide concentration may be higher or lower than the estimated value, depending on the nature of the variance. If the field environmental conditions differ from the assumed conditions, but are accurately known from independent information, then the calibrated estimates may be effectively corrected. If the field environmental conditions are unknown, the concentration estimate is subject to irreducible uncertainty; such is the case with the measurements in the SDA.

The suggestion has been made that exhumed waste could be used to construct calibration models that would more closely represent the subsurface environmental conditions at the SDA. The waste would be placed in drums and arranged around the logging tool for calibration measurements. This method would produce new values for K and F in Equation (2). Multiple alternative configurations of the waste could be modeled, resulting in multiple values for K and F; however, the specific subsurface waste zone conditions remain an unknown, and selection of the appropriate K and F values would not be possible.

3.1 Existing Logging-Tool Calibration

The passive spectral gamma-logging tool used for Type A probehole logging at the SDA was calibrated using standards constructed from concrete loaded with various, known, uniform concentrations of potassium, uranium, and thorium. The uranium and thorium in the calibration models were determined to be in secular equilibrium with the uranium and thorium decay products. The calibration models are steel tank enclosures with alternating zones of blank and loaded concrete. Each tank has two enriched zones 1.2 m (4 ft) high that are separated by a 1.5-m (5-ft) thick blank zone. Blank zones 91 cm (3 ft) thick also are located above the upper enriched zone and below the lower enriched zone. During calibration, multiple measurements are made with the detector centered in the enriched zones of each model. This procedure can be used to define the tool count-rate dependence on radionuclide concentration under the following conditions:[d]

- Radionuclides uniformly distributed around the probehole
- Probehole diameter equals 15.2 cm (6 in.)
- No probehole casing
- Air-filled probehole
- Formation bulk density equals 1.82 to 1.92 g/cc
- Potassium, uranium, and thorium only.

This calibration has very limited application because of the highly restrictive conditions. Table 1 compares the calibration conditions to conditions expected at the SDA. Only Condition 3 of Table 1 has an exact correspondence between calibration and field conditions. For Conditions 1, 2, and 4, the variance between calibration and field conditions is fully known and it is possible to make accurate corrections. Probehole diameter and casing corrections are made by adjusting the observed count rates to compensate for the difference between calibration and field conditions. These adjustments are determined from experimental data collected under controlled conditions or from detailed mathematical simulations. Corrections for different radionuclides are made by establishing an efficiency hnction that accounts for differences in the logging tool response at different gamma-ray energies associated with each radionuclide of interest.

d. Additional details about construction of the passive spectral-gamma calibration standards are contained in Stromswold (1994) and Koizumi (1994).

Table 1. Comparison between calibration conditions and actual conditions for Subsurface Disposal Area logging.

Condition Number	Calibration Condition	Subsurface Disposal Area Condition
1	Probehole diameter equals 15.2 cm (6 in.)	Probehole diameter equals 14 cm (5.5 in.)
2	No probehole casing	12.7 cm (0.5 in.) steel casing.
3	Air-filled probehole	Air-filled probehole.
4	Potassium, uranium, thorium only	Potassium, uranium, thorium, plutonium, americium, neptunium, cesium, cobalt, and others.
5	Radionuclides uniformly distributed	Radionuclides may be locally uniform or highly discontinuous.
6	Formation bulk density is uniform and equals 1.82 to 1.92 g/cc	Formation bulk density may be locally uniform or highly discontinuous in the range up to 11.4 g/cc (density of lead) with typical undisturbed soil bulk density in the range from 1.4 to 1.9 g/cc.

The variance between calibration and field conditions for Conditions 5 and 6 is variable and unpredictable and no accurate correction is currently available. Field conditions will occasionally correspond with calibration conditions on occasion; however, it is impossible to recognize when this occurs because there is no independent means for measuring these conditions.[e] Given the known heterogeneous character of SDA waste materials, it is evident that no distribution of these parameters can be considered as a standard. In this case, calibration assumptions for radionuclide and density distribution are chosen purely on the basis of mathematical and operational convenience.[f] Thus, two additional assumptions are made during the SDA logging program calibration process:

- Homogeneous radionuclide distribution
- Homogeneous matrix density.

These assumptions are entirely appropriate because they (1) can be supported using an existing calibration facility and (2) create a reproducible measurement standard that is independent of tool hardware. Alternatively, these assumptions are clearly unrealistic representations of field conditions, which affects their quantitative validity.

3.2 Existing Calibration Limitations

Calibrations performed for logging tools, both in the oil and gas industry (see Appendix A) and for logging the Type A probes in the SDA, require varying amounts of rigor accompanied by multiple quality assurance and quality control checks to verify and validate accuracy of the calibrations. This rigor assures that the logging tool will yield correct and reproducible results when calibration assumptions are met and

e. For radionuclides that emit multiple gamma-ray energies, uniformity can theoretically be assessed by evaluating gamma-ray attenuation as a function of gamma-ray energy. This method has been proposed (Jewell, Reber, Hertzog 2002, Attachment A), but has never been formally developed. Formation bulk density is commonly measured using a gamma-gamma density logging tool, but these tools are not designed to work through casing because of the severe attenuation of the gamma-ray source.

f. Oil, gas, and mining downhole nuclear logging applications are frequently, but not always, based on the assumption of uniform conditions.

allows comparison between measurements made at different times or using different tools. However, these calibrations are not without limitations. Limitations associated with calibrations of logging tools used in the oil and gas industry are relatively few. The limitations arise from inevitable deviations that occur between assumptions made during the calibration processes and actual conditions present in the field during logging operations. The limitations are minimized by the fact that the logging environment in the oil and gas industry is, for the most part, homogeneous. Heterogeneity in the subsurface environment for oil and gas exploration are typically introduced during the drilling process; as such, they are easily quantified and accounted for. Examples include mudcake, drilling mud, and casing or drill steel. Other heterogeneity associated with the lithology are also relatively easily quantified through the use of multiple types of borehole measurements. However, heterogeneity in the lithology typically occurs over a relatively large scale (i.e., on the order of several feet), and sharp lateral discontinuities are rare.

In stark contrast to the oil and gas industry, logging activities associated within the SDA waste zone must accommodate a very high degree of heterogeneity. Both vertical and lateral discontinuity is commonplace. Primary limitations for Type A probehole logging arise from the blanket assumption that property being measured is uniformly distributed in an infinite, homogeneous, soil matrix. It is well known and accepted that this condition does not exist in the subsurface waste zone in the SDA. Heterogeneity arises from the diversity of known waste types and waste disposal practices. As a result, the absolute concentrations associated with passive spectral gamma logging are not accurate depictions of the true concentrations of radiological contaminants in the subsurface. Furthermore, no method is known for obtaining accurate independent information about radionuclide distribution and matrix density that could hrther reduce the uncertainty.

3.3 Probehole P9-20 Modeling

A detailed investigation of the mass of Pu-239 near Probehole P9-20 was conducted while recognizing the limitations of initial calibrations used for logging tools deployed in the SDA; specifically, the passive spectral gamma tool. The investigation involved placement and logging of a cluster of probes surrounding Probehole P9-20. Two separate modeling exercises were performed to estimate the Pu-239 mass distributed within the probe cluster (Jewell, Reber, and Hertzog 2002). Both methods allowed simulation of nonhomogenous matrix and radionuclide distribution. These modeling exercises clearly demonstrated the interdependence between density distribution, mass distribution, and the amount of Pu-239 mass. Without substantive information with which to fix these interdependent parameters, the Pu-239 mass estimate obtained by modeling was essentially unbounded. However, by fixing these parameters within conservative density limits for a range of worst-case mass distribution scenarios, modeling showed that the Pu-239 mass was unlikely to be less than 319 g and unlikely to be greater than 2,217 g. The worst-case mass distribution scenarios incorporated the following conservative assumptions:

- Single, compact source
- No surrounding void space
- Self shielding correction of 20% (1,000 μm plutonium particle size).

The estimated plutonium mass would be substantially lower if multiple, distributed plutonium particles (i.e., no significant self absorption) were assumed rather than a single, compact mass.

The lower plutonium mass limit was based on assumed presence of a low-density waste and soil matrix in the Probehole P9-20 vicinity. The higher plutonium mass limit was based on reasonable maximum values for density of the waste and soil matrix. Although providing no definitive answer, this result proved to be more usehl for planning purposes than the standard calibration and suggested an alternative calibration approach for the passive spectral gamma-ray logging tool.

4. ALTERNATIVE TYPE A PROBE PASSIVE GAMMA-RAY LOGGING CALIBRATION METHOD AND SYSTEMATIC DATA ANALYSIS

Because of the critical influence of matrix density and radionuclide mass distribution assumptions, the existing calibration method cannot be relied on to provide usehl quantitative radionuclide mass estimates. In view of this significant limitation, it is believed that the best possible calibration method should simply provide bounding limits on radionuclide mass. These bounding limits are established by setting bounding conditions on matrix density and radionuclide distribution as part of the calibration assumptions. An alternative calibration objective may be stated as the desire to use Type A probing data to differentiate between TRU (greater than 100 nCi/g) and non-TRU (less than 100 nCi/g) waste. Developing the alternative method to achieve the objective included the following:

- Determining limiting conditions of possible source geometries and waste matrix density
- Modeling source geometry to determine required minimum probe spacing (or maximum spacing density)
- Developing methodology for data collection and analysis that results in classifying the subsurface as non-TRU, possible-TRU, or definite-TRU.

Each of these items are discussed in hrther detail in the following sections and include assumptions and limitations associated with each aspect of the calibration.

4.1 Source Geometry and Matrix-Density Bounding Conditions

Defining source geometry and matrix density are essential elements for developing a calibration method for logging measurement systems. These bounding conditions are typically governed by the most influential variables in the system, which in the case of the borehole measurements with the passive spectral gamma system is the source distribution and the waste zone density.

The objective of identifying waste material that exceeds 100-nCi/g TRU content cannot be accomplished because of known heterogeneous conditions in the SDA; therefore, it is necessary to first modify this objective to achieve results with practical utility, by using Type A probing data to identify waste material that could exceed 100 nCi/g TRU content.

As such, two source and probe geometries were identified as limiting conditions that would likely be encountered in the SDA:

1. A point source of Pu-239 centered inside a 55-gal drum of waste, centered inside a triangular grid defined by three close-spaced probes

    a. The common waste material disposed of in the SDA surrounding the point-source TRU is roaster oxide waste with an average density of 1.7 g/cm^3.

2. A distributed Pu-239 source inside a 55-gal drum of waste, centered inside a triangular grid defined by three close-spaced probes. These source geometries are depicted in Figure 4. Based on these two scenarios, the following assumption was made about the source and waste matrix:

    a. Total Pu-239 activity associated with a 55-gal drum of roaster oxide with a concentration of 100 nCi/g is 3.629×10^7 nCi, which relates to a mass of 0.58 g of Pu-239.

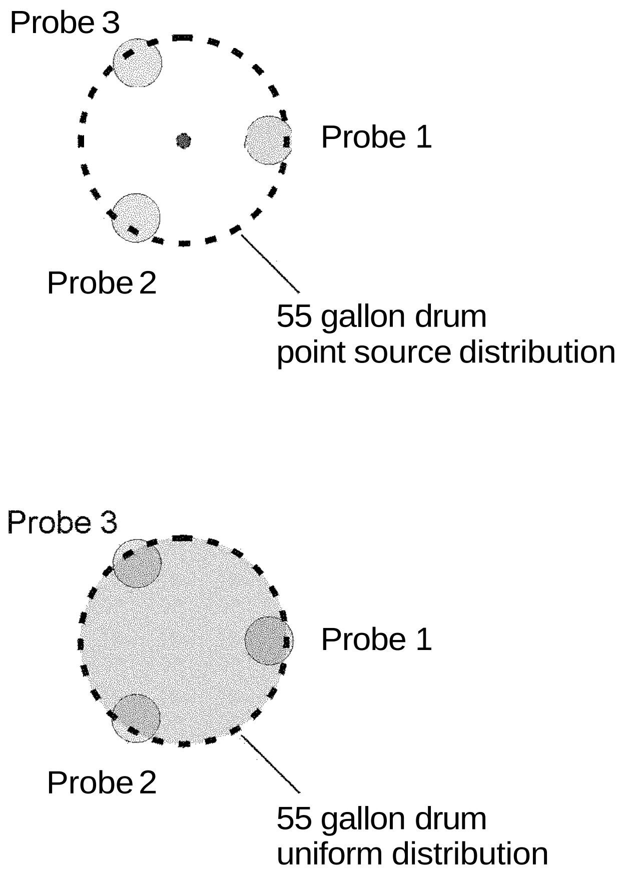

Figure 4. Schematic of point-source and distributed-source geometries for gamma-ray tool calibration.

The next step in establishing the calibration is to determine the probe-spacing requirements. The approach was modeling. It can be shown through a simple modeling exercise that the weakest signal at the detector is from the point-source geometry. Because gamma rays have limited capacity to penetrate the surrounding medium, it is important that at least one probe occurs within the zone of detectable gamma-ray flux that surrounds the hypothetical source.

The required probe spacing is determined by using the point source model to analyze Pu-239 source mass at Probehole P9-20. The source size was set at 0.58 g and the probe locations were adjusted until the observed count rate matched the estimated lower limit of detection as determined from previous SDA logging. For a 200-second count using a 35% HPGe detector, the lower limit of detection for Pu-239 was approximately 30 nCi/g, which corresponds to a count rate of 0.43 counts/second. Disregarding self-absorption in the 0.58-g Pu-239 source, the maximum probe spacing to detect the source is 0.5 m (1.8 ft).

4.2 Passive Spectral Gamma Data Collection and Analysis

A systematic, robust approach to the analysis of probing data is key to providing meaningful quantitative results. This section outlines the basic approach to the data analysis to be used in conjunction with the alternative calibration. The actual data analysis methodologies outlined here have not yet been developed; however, the methodologies presented are based on sound, scientific principles.

Based on the bounding conditions for the source geometry presented above, the maximum probe spacing will be 0.5 m (1.8 ft), resulting in the installation of an estimated 15,600 probes/acre. These Type A probes then will be logged using the passive spectral gamma tool to identify the presence or absence of TRU-containing waste in the subsurface. In the first screening step, areas where no TRU radionuclides are detected may be immediately classified as non-TRU. The remainder of the subsurface is tentatively classified as possible-TRU.

In the second processing step, spectral shape analysis is conducted to determine if a uniform radionuclide and density distribution model can be justified. If so, some of the possible-TRU areas probably can be reclassified as non-TRU.

In the third processing step, the remaining possible-TRU areas are subjected to rigorous Monte Carlo N-Particle Transport Code System (LANL 1997) modeling that will simultaneously solve for radionuclide and density distribution within the area of interest. Multiple model solutions will be possible, but in some cases all solutions will point to non-TRU or definite-TRU conditions, in which case a definitive classification can be made. The flow chart in Figure 5 shows the proposed systematic data analysis.

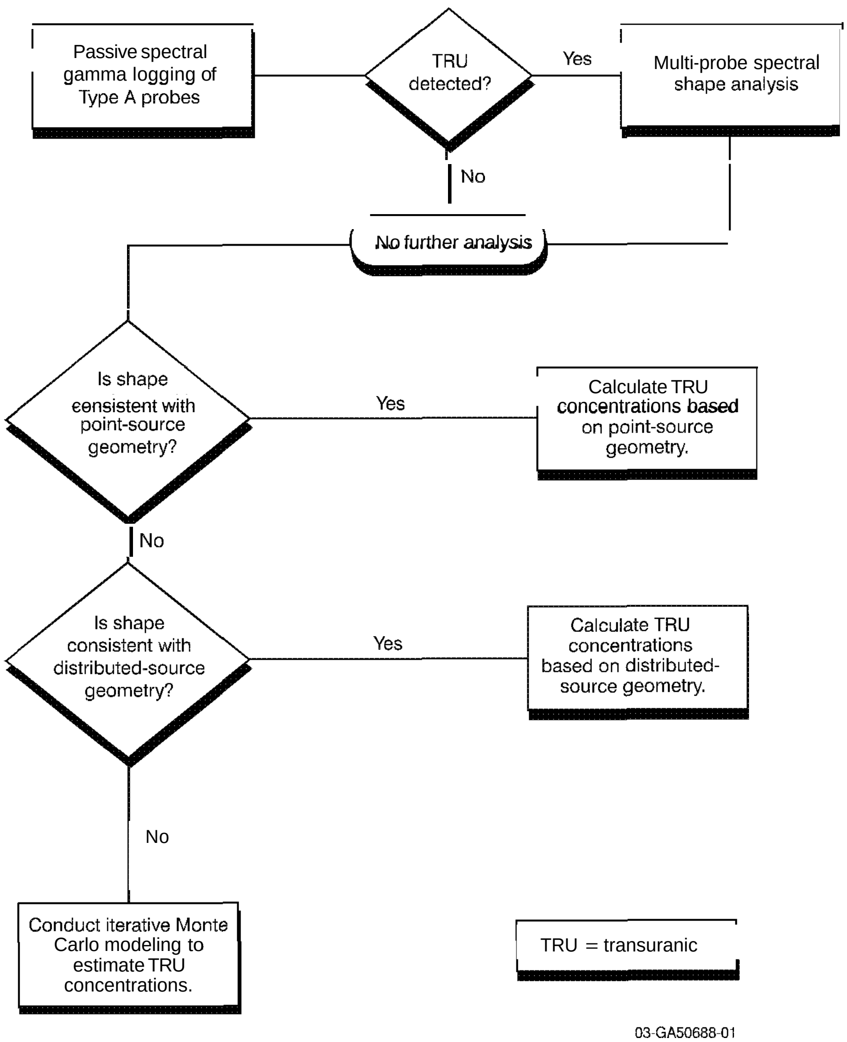

Figure 5. Systematic passive gamma spectral analysis for transuranic contaminants in the Subsurface Disposal Area.

4.3 Limitations of Alternative Calibration and Data Analysis Methods for Passive Spectral Gamma Logging

Proposed calibration and processing methods described in this document will provide a defensible basis for calculating TRU content in the SDA subsurface; however, these methods also have serious limitations. As with existing calibration, limitations arise from discrepancies between assumed and actual conditions that exist in the waste zone within the SDA. An error caused by wrong assumptions may be categorized as a false-negative (i.e., scenarios that would cause TRU waste to go undetected). This results in failure to detect TRU waste, or a false-positive (i.e., scenarios that would cause non-TRU waste to be incorrectly classified as TRU), which results in non-TRU waste being incorrectly classified as TRU. Tables 2 and 3 list scenarios that would produce these types of errors.

Table 2. False-negative scenarios.

Scenario	Description	Probabilitv	Corrective Action
FN1	TRU material consists of Np-237 rather than Pu-239.	High	Closer probe spacing required
FN2	TRU surrounded or intermixed with high-density waste or matrix material (e.g., lead or steel).	Medium	None—no known method to independently determine density.
FN3	TRU particles greater than about 100 µm, resulting in significant self-absorption effect.	Low	None—no known method to independently determine particle size.

TRU = transuranic

Table 3. False-positive scenarios.

Scenario	Description	Probability	Corrective Action
FP 1	TRU material consists of Am-241 rather than Pu-239.	High	Correct for difference between americium vs. pltuonium specific activity.
FP2	TRU surrounded or intermixed with low-density waste or matrix material (e.g., plastic or void space).	Very high	None—no known method to independently determine density.

TRU = transuranic

Tables 2 and 3 may be considered a performance measure of the calibration and processing scheme. Although this level of performance is only achieved using a very dense probe-spacing pattern, Scenario FN1 indicates that TRU radionuclides having lower specific activity than Pu-239 may still be missed unless even more probes are installed.

Scenarios FN2 and FP2 measure the validity of the assumed waste- and soil-matrix density. By assuming a high density, the method produces fewer false-negatives but is extremely vulnerable to false positives. The proportion of false negatives to false positives maybe changed by assuming a different density, but the total number of errors cannot be decreased in any way without independent density information. In one limit, false negatives may be completely eliminated by assuming a lead waste matrix (density = 11.4g/cc). This new density assumption would require a denser probe spacing pattern and would result in an excessive number of false positives. In the other limit, by assuming a low-density void-filled matrix, it is possible to virtually eliminate false-positive errors, but at the expense of creating an excessive number of false negatives. The fundamental ambiguity caused by unknown density is simply irreconcilable.

Scenario FN3, although relatively unlikely to occur, illustrates another irreconcilable limitation of any gamma-ray-based method for classifying TRU. As a TRU particle becomes physically larger than about 100 μm, it begins to reduce gamma-ray emissions through self-absorption. At some point, a maximum gamma-ray emission level is achieved and never exceeded no matter how large the TRU particle becomes (i.e., above a certain particle size, gamma-ray flux is no longer sensitive to the amount of TRU).

5. ALTERNATIVE MEASUREMENT SYSTEMS

Effective characterization of subsurface soils and buried waste at the SDA, using a near real-time in situ field survey system to locate and determine the distribution of TRU contamination, would be valuable in guiding environmental restoration activities. Therefore, an effort was undertaken to determine whether reliable surface monitoring systems are available to detect subsurface TRU elements (plutonium in particular) and identify TRU hot spots to diagnose the need for remediation at the SDA.

Useful tools should provide data with good spatial resolution with short data-acquisition times at low cost. An important criterion is that the technology also must be capable of providing depth profiles in conditions at the SDA, where the average depth of buried waste is 2.7 to 3 m (9 to 10 ft) and the clean soil overburden ranges from 0.9 to 2.4 m (3 to 8 ft).

Several surface-monitoring systems were identified for evaluation:

- Long-range alpha detector
- Long-range neutron detection
- Prompt gamma–neutron-activation-analysis technology.

These technologies and the feasibility of their use in the SDA for identifying TRU in the subsurface are discussed in Appendix B. Additionally, new logging tools are being developed by the nuclear measurements group at the INEEL including prompt-fission-neutron and pulsed-neutron gamma-ray spectrometry tools. These tools also are described in Appendix B.

6. CONCLUSIONS AND RECOMMENDATIONS

Known heterogeneous conditions in the SDA subsurface create the likelihood that most, if not all, Type A probeholes violate the spectral gamma-ray logging tool calibration conditions that assume homogenous radionuclide distribution and homogenous density of the waste and soil matrix. Recalibration of the spectral gamma-ray tool to a new standard condition is not recommended because no meaningful standard exists for radionuclide distribution or waste and soil matrix density. Furthermore, information concerning radionuclide distribution and waste and soil matrix density cannot be obtained by any known independent measurement method. Consequently, the spectral gamma-ray measurements cannot be meaningfully corrected for heterogeneity.

An alternative approach to calibration and processing of Type A logging data was developed with the sole purpose of screening the subsurface for non-TRU waste. To implement this method, a set of standard conditions is defined under which a subsurface volume surrounded by probes may be categorized as either non-TRU or possible-TRU. Possible-TRU areas are subjected to additional processing steps by which some may be recategorized as non-TRU.

By the proposed method, subdivision of the entire SDA into non-TRU and possible-TRU subvolumes can be achieved. However, the method requires the installation, logging, and analysis of approximately 15,600 Type A probes per acre. Furthermore, both false-positive errors (non-TRU waste categorized as possible-TRU) and false-negative errors (TRU waste categorized as non-TRU) are possible. The proportion of the two error types depends on the choice of standard conditions. For example, if standard conditions are chosen conservatively, false-negatives mostly may be avoided at the expense of a large number of false-positives.

Alternative radiation measurement technologies were investigated to determine their applicability for differentiating TRU waste. Several large-area, long-count-time detectors were considered for deployment on the SDA ground surface. As a class, these tools were found to have no utility at the SDA because of the limited penetration of alpha particles, gamma rays, and neutrons through soil. New emerging radiation logging tools may be very usehl for TRU differentiation if employed in a high probe-density survey as depicted in this report. The primary benefit of these tools will be as additional constraints on three-dimensional modeling. These tools will not significantly improve the ability to directly identify non-TRU waste because they will suffer from the same uncertainties about source geometry and soil and waste matrix density.

Despite the difficulties (improbabilities) associated with calibrating the tools for quantitative purposes, the existing data set and any logging data gathered in the future may be used qualitatively in identifying waste types and waste zone composition and in providing information directing quantitative investigations.

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Appendix A

Logging Tools and Calibrations in the Oil and Gas Industry

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Logging Tools and Calibrations in the Oil and Gas Industry

INTRODUCTION

In 1927, C. and M. Schlumberger made the first subsurface geophysical measurements near Paris with an electrical current experiment that used an electrode placed at a series of horizontal positions on the surface of the ground to detect variations in the geological structure below the surface. During the 1930s, these experiments evolved into a simple tool consisting of an electrode and a current source that performed continuous measurements of the subsurface resistivity by lowering the tool into a borehole. The interpretation of these resistivity logs for oil exploration helped to determine bed thickness, identify clay-rich and hydrocarbon-bearing zones, and provide a rough estimate of formation permeability. All these qualities could be correlated between holes that provided a rough map of the oil and gas reservoirs in the subsurface. Since then, the critical need for borehole measurements in the oil and gas exploration and production industry has lead to the development of a wide variety of highly sophisticated and expensive downhole measurement tools including electrical, nuclear, sonic, and acoustic logging instruments.

The following is a summary of common tools used in the logging industry, and the calibration efforts expended to ensure accurate down-hole measurements. A more detailed description of the logging tools and the extensive calibration methodologies used in the oil and gas industry is contained in the following subsection.

A typical suite of down-hole tools are used during the exploration for oil and gas deposits. These tools include the following:

- Density tools—These tools comprise a Cs-137 source and two detectors (one near the source and another hrther away) and are used to obtain accurate measurements of the formation density. The data also are used to calculate formation porosity.

- Neutron porosity tools—These tools comprise a neutron source (either chemical- or accelerator-produced neutrons) and one or more neutron detectors and are used in conjunction with density measurement data to measure the formation porosity, which is directly related to the volume of petroleum product.

- Passive gamma-ray tools—Passive gamma-ray tools comprise single or multiple gamma-ray detectors and are used to measure the natural gamma-ray flux in the subsurface and as indicators of hydrocarbon-bearing zones.

- Active neutron tools—Active neutron tools comprise a neutron source (either chemical- or accelerator-produced neutrons) and one or more detectors. The detectors may be either neutron or gamma-ray detectors that measure induced responses in elements in the formation. These tools are used to measure physical properties of the formation (e.g., water saturation content) and to characterize geological sequences and phenomena.

It is important to note that though multiple tools are used for exploration of oil and gas, the independent measurements made with each tool are not mutually exclusive. The complex data processing activities use and combine data from independent measurements to produce useable information about the subsurface.

Logging Tools

Density Tools

In saturated formations, density measurements are used to determine porosity. If the formation grain density is known, then porosity can be calculated from the density log. Alternatively, porosity and density logs can be used together to calculate grain density. Bulk density information can be combined with seismic velocity to generate synthetic seismograms. In combination with the neutron log, the density allows definition of lithology and lithologic boundaries.

A reliable density measurement requires good contact between the tool pad (or skid) and the formation (borehole wall). In litho-density tools (LDT), the presence of mudcake and hole irregularities is automatically corrected using a spine-and-ribs chart or algorithm based on an extensive series of laboratory calibration measurements. The spine is the locus of the two counting rates from the near and far detectors without mudcake, and the ribs trace out the counting rates for the presence of mudcake at a fixed formation density. The near- and far-spaced detector count rates are automatically plotted on this chart and corrected for departure from the true value.

Litho-Density Tool. A typical example of a density tool used in the oil and gas industry is the LDT provided by Schlumberger. This tool consists of a Cs-137 source (approximate strength is 1.5 Ci) and two detectors (one near the source and another hrther away) mounted on a shielded skid that is pressed against the formation by a hydraulically activated eccentering arm. The 662-KeV gamma rays emitted by the source into the formation interact with atomic electrons in the formation elements by (1) Compton scattering and (2) photoelectric absorption.

Compton scattering is an elastic collision by which energy is transferred from the gamma ray to electrons in the formation. This interaction forms the basis of the density measurement. Because the number of scattered gamma rays reaching detectors is directly related to the number of electrons in the formation, the tool responds to the electron density of the formation rocks (and pore fluids), which is, in turn, related to the formation bulk density.

Photoelectric absorption occurs when the gamma rays reach low energies after being repeatedly Compton-scattered by electrons in the formation. The photoelectric effect index, P_e, is determined by comparing the far detector high-energy counts (where only Compton scattering occurs), with low-energy count rates (where the count rates depend on both interactions). The far detector is used because it has a greater investigation depth. The response of the near detector, which is influenced more by rough or irregular borehole surfaces, is used to correct the density measurement for these effects.

Neutron Porosity Tools

In reservoir engineering for the oil and gas industry, porosity is one of the most important logging measurements, besides oil saturation. Porosity defines the volume of the formation in which fluids reside. With a measurement of oil or, conversely, water saturation, the total hydrocarbon volume can be assessed. Two of the most commonly used neutron porosity tools in the oil and gas industry are the dual porosity compensated neutron tool (CNT-G) and the accelerator porosity tool (APT).

Dual Porosity Compensated Neutron Tool. The CNT-G uses an americium/beryllium chemical neutron source that emits fast neutrons. These fast neutrons are scattered and moderated by collisions with nuclei in the formation. Hydrogen is the most efficient moderator, and in the presence of large amounts of hydrogen (water or hydro-carbons) the fast neutrons are efficiently and quickly slowed to thermal energies. In the CNT-G, two pairs of detectors measure both epithermal (intermediate energies) and thermal (slow) neutrons. The result is a thermal and epithermal neutron-derived porosity and, therefore, dual-porosity logging. Thermal porosity is susceptible to the effects of thermal absorbers in the formation, such as chlorine and other elements with large thermal cross sections (e.g., boron, rare earths, and transuranic [TRU] waste materials). The epithermal porosity is used to detect when such absorbers may be affecting the thermal porosity readings. Although epithermal-neutron porosity measurements are, in principle, superior to thermal porosity measurements, the detector spacings in the CNT-G were selected to maintain sufficient count rates rather than optimum porosity accuracy under a wide range of environments. For that reason, an accelerator-based epithermal neutron porosity tool, such as the APT (discussed below), is preferred.

Accelerator Porosity Tool. The APT uses a powerful pulsed-electronic neutron source instead of a chemical source. The large neutron source yield (3×10^8 n/second) allows epithermal neutron measurements and detector shielding, resulting in porosity values that are less influenced by environmental conditions. Five detectors (four epithermal and one thermal) provide information for porosity (or moisture in unsaturated formations), gas detection, shale evaluation, improved depth resolution, and borehole corrections. Epithermal neutron measurements are much less influenced by changes in lithology (e.g., those caused by shales, limestone, or dolomites), and unaffected by large changes in the formation macroscopic neutron absorption cross-section caused by saline water in oil reservoirs, or by chlorinated organics (e.g., CCl_4) and TRU in buried waste. In addition, the pulsed neutron source provides for two new measurements: (1) an epithermal slowing down time that is used to evaluate tool standoff from the borehole wall and to correct the porosity measurement and (2) a formation thermal neutron cross-section (sigma) that is a usehl indicator for the presence of elements of high thermal neutron-capture cross-section (e.g., boron, chlorine, rare earth elements commonly associated with clays in shales, and possibly TRU waste). The APT epithermal porosity measurement is corrected for borehole conditions (e.g., size, salinity, mudcake, and tool standoff) and is nearly, though not entirely, independent of lithology effects.

Passive Gamma-Ray and Spectral Measurements

The natural gamma-ray log measures the total radioactivity in the subsurface from potassium, uranium, and thorium. This basic measurement, though unsophisticated, has been used extensively in mining and the oil and gas industry to locate zones of high radioactivity usually associated with shale beds. Geologically, shale is important because it usually is a permeability barrier and provides a trap for hydrocarbons. In the right geological setting, highly radioactive zones also can indicate regions containing high uranium-ore concentrations.

Elaborate interpretation schemes have been developed for passive gamma-ray (potassium, uranium, and thorium) measurements to distinguish feldspar, micas, and clay minerals in sediments. Because the results often are ambiguous, the ratio of radioactive elements is compared with the photoelectric effect index from the LDT in some interpretation schemes. However, advanced interpretation methodologies have been developed where passive spectral measurements are combined with other neutron-induced spectral logs (discussed below) to determine volumetric analysis of complex mineral mixtures leading to detailed subsurface geochemical analyses.

Natural Gamma Tool. The natural gamma tool uses a large sodium-iodide scintillation detector to measure the natural gamma-ray radiation of the formation and a five-window spectrometer to resolve the detected gamma-ray energy spectrum into the three components of naturally occurring radiation: potassium, uranium, and thorium. The depth of investigation depends on several factors: hole size, mud density, and energy of the gamma rays. The natural gamma tool also is affected by the presence of bentonite, or potassium-chloride (KCl) in the mud, and special corrections must be used to correct for the KCl-mud contribution to the potassium measurement.

Neutron-Induced Spectral Measurements

Thermal Decay Time Measurements. Since the 1960s, thermal neutron decay-time logging is used extensively in the oil and gas industry for water saturation analysis through discrimination between saline water and hydrocarbons. The basic thermal decay time measurement determines the thermal neutron absorption decay rate in the formation, which is a measure of the formation neutron absorption or capture cross section.

The formation neutron capture cross section depends directly on the volumetric mixing of low neutron capture cross section of the rock minerals and higher capture cross sections of the fluids. Combined with other measurements (e.g., neutron porosity, gamma-ray, and tool-detector array count rate ratios) the thermal neutron decay logs can be used to determine apparent water salinity, formation porosity, shale content, and gas in the formation.

Prompt and Delay Fission Neutron Tools. Prompt fission neutron (PFN) and delayed fission neutron (DFN) devices were developed in the mid-1970s for uranium exploration and mine planning. They also have been evaluated and used to detect and quantify spilled or buried fissile materials inside or leaking from subsurface containers at weapons complex sites. These highly sensitive devices use a pulsed neutron source, or isotopic sources such as Cf-252 to irradiate fissile materials, causing them to undergo neutron-induced fission. The fission process releases secondary neutrons, and the fissile material in the vicinity of the probe is quantified by measuring the secondary neutrons. The PFN measurement is the most sensitive and depends on the ability to time-pulse a neutron generator on and off completely so that secondary fissile neutrons can be detected (in a background-free time gate) between bursts of source neutrons from the generator. The DFN techniques employ detection of the delayed neutrons from fission that persist long after the source of neutrons has been removed. The DFN technique, although less sensitive than PFN, has one advantage in that it can use either isotopic sources or a pulsed neutron source. These devices can then be used to sensitively measure transuranic elements.

Fast Neutron-Scattering Induced Spectroscopy. Fast neutron-scattering induced spectral measurements for carbon to oxygen (C/O ratios) were introduced in the oil and gas industry in the mid-1970s for determining oil saturation in complex environments where thermal neutron decay time measurements would not work, such as fresh water (no cross section contrast between oil and water) or unknown water salinity. Carbon concentration is a direct measure of hydrocarbons in the formation.

Gamma-Ray Spectrometry Tool. The gamma-ray spectrometry (GST) operates in two timing or spectrometer modes: (1) inelastic neutron scattering, which is used to measure carbon and oxygen; and (2) the capture-tau mode, which employs prompt neutron capture reactions to measure elemental concentrations for many other elements including silicon, calcium, sulfur, iron, titanium, and gadolinium. The capture-tau mode also results in a measurement of sigma, the macroscopic neutron cross-section of the formation. The GST neutron source is a pulsed tritium source that bombards the formation with 14 MeV neutrons. Through inelastic and elastic neutron-scattering reactions with the atoms in the formation, the neutrons progressively lose energy until they reach a thermal equilibrium with the formation. The thermal neutrons are captured by elemental nuclei in the rock and, when this occurs, the nuclei emit a gamma-ray at a unique energy, which is characteristic for each capture element (e.g., silicon, calcium, iron, and others). Inelastic scattering of fast neutrons also induces the bombarded nuclei (e.g., carbon and oxygen) to emit signature gamma rays that are used to identify their presence and measure their concentrations. The emitted gamma rays are captured by a large sodium iodide detector in the GST and measured by multiple 256-channel analyzers to separate the inelastic spectra measured during the fast-neutron burst from the capture and background spectra measured between bursts. Spectral processing is based on a weighted-least-squares unfolding of a large number of elemental standards from the observed spectra, including those from carbon, oxygen, silicon, calcium, iron, titanium, sulfur, hydrogen, and chlorine.

Geochemical Logging Tool. The gamma-logging tool (GLT) uses three separate modes of gamma-ray spectroscopy to obtain measurements of most of the major oxides, which make up sedimentary and igneous rocks. Initial measurements provide estimates of silicon, aluminum, iron, calcium, potassium, sulfur, uranium, titanium, gadolinium, and thorium (together with hydrogen and chlorine). Estimates on sodium, magnesium, or manganese and other elements are possible with secondary processing. The GLT provides gross geochemical information about the formation, which is particularly usehl when combined with other logs. The data can be used directly for characterization of geological sequences and phenomena, and are excellent for geotechnical zoning. The GLT consists of four components:

- An NGT, which measures potassium, uranium, and thorium
- Aluminum activation clay tool (AACT), which is a carrier for a low-energy californium
- A Cf-252 neutron source and a second NGT that is used to detect formation aluminum activated by the Cf-252 neutron source
- A GST used to detect all the other elements in pulsed-neutron-capture-mode spectroscopy measurements.

Californium is used instead of the conventional Am/Be neutron source because its neutron energy spectrum has a lower average energy (2 MeV instead of 4.5 MeV), thus reducing fast neutron reactions (e.g., Si(n,p)Al), which interfere with the aluminum measurement.

Logging Tool Calibration

Tools developed for the oil and gas industry are required to be accurate, often to within less than 1% absolute error. Commercial tool calibrations are necessary and expected for every measurement. Typically, all nuclear tools are required to have three classes of calibrations:

1. Primary tool calibrations, which include environmental corrections
2. Shop calibrations
3. Well-site calibrations.

Primary Tool Calibration

Primary tool calibration is the highest-level calibration and defines the overall tool response (e.g., count rates vs. porosity) in a wide range of formation environments. This is done at the tool engineering or manufacturing center when the tools are designed and built. These calibrations involve measuring tool responses under the normal operating conditions (i.e., borehole sizes and borehole environments [fluids and mudcake]), and over the expected range of the principal measurement property (e.g., range of formation density or porosity the tool is designed to measure). Calibration formations consist of

- Precision blocks of known-density material about 91 cm (3 ft) high by 60 cm (2 ft) wide and approximately 30 to 60 cm (1 to 2 ft) thick

- Large, carefully constructed tanks 1.8 m (6 ft) high and 1.2 to 1.5 m (4 to 5 ft) in diameter, filled with the formation media (porous sand) to a designed porosity.

Calibration models are constructed with a tool access hole that penetrates through the center of the models, simulating a borehole. Typically, calibration tanks are designed such that the formation fluid can be exchanged for another of different properties (e.g., replacing saline water with fresh water). Quite often, the porous formations are filled with different hydrocarbon mixes to simulate different oil and water saturation ratios.

The bulk formation under investigation is usually homogeneous, but the environment surrounding and between the tool and the formation is not. Therefore, other environmental (borehole and formation) properties can vary and affect the tool response, such as those listed below:

- Changing thickness of mudcake in the borehole
- Borehole fluid variations
- Subsurface temperature changes
- Pressure changes
- Changes in neutron absorption
- Density changes
- Variations in tool standoff (or position in the borehole)
- Varying quantities of naturally occurring radioactive material in the mud that are different from the formation.

These environmental effects are always investigated during this stage of tool calibration to understand their effect on tool response and to develop environmental correction algorithms for these effects on the primary response. This phase of the primary calibration process is often called environmental correction calibrations, and the logging companies have built large laboratories to provide the extensive range of test tanks and formations to perform these calibrations. For example, the Schlumberger Environmental Effects Calibrations Facility is a 60,000-ft^2 laboratory built in the early 1980s at a cost of over *$3* million and dedicated exclusively for calibrating nuclear logging tools used for oil and gas exploration. It is important to note that although the formations under investigation are, for the most part, homogeneous, the heterogeneity that may be encountered in the field is relatively easily defined and bounded. As such, correction factors for these conditions are measurable.

Based on this process, the logging service companies can transform the tool count-rate responses to primary measurement objectives (e.g., density, porosity, element concentration, or some other physical property) using the algorithms derived from the primary calibration and environmental correction data. This is the only way to ensure accurate tool measurements under a wide variety of environments experienced during downhole logging.

Shop Calibration

The second class of calibration, called the shop calibration, is a one- or two-point primary response calibration done with a test tank or test block. Shop calibration simulates actual operating conditions and is performed at the regional shop location where the tools are located and ready for field use. Often, the shop test tanks are precision duplicates of the primary tool-calibration tanks. Shop calibrations are performed before and after the tool is taken to a well site for a logging operation.

Well-site Calibration

The final class of calibration, the well-site calibration, is done at the well site just before the logging operation commences. This is typically more of an instrument check as opposed to a tool calibration, because no room is available at the well site for a large tank or calibration block. Well-site calibrations are completed using a known standard and verifying the tool response is within acceptable limits.

Specific Tool Calibrations

Density Calibrations. Primary tool calibrations for the density tools involve a wide range of formation densities ranging from dense, zero-porosity formations to lighter, high-porosity formations. For example, Schlumberger calibration of the LDT involves basic calibrations for the tool response with six different weights and three thickness of mudcake in over 75 primary calibration measurements. At least 37 calibrations are used just to develop the mud-cake correction algorithm. Nine calibration blocks are used to develop the primary photoelectric response, including marble-, dolomite-, sulfur-, aluminum-, and quartz-sand-blocks along with water-tank measurements. Shop tests include the use of two or three test-block calibrations (e.g., aluminum and sulhr blocks). Well-site tests include count-rate measurements inside the casing before the tool is lowered fully into the well. A repeat pass of one section of the well (typically 15 to 30 m [50 to 100 ft]) serves as a quality assurance and quality control check to compare logging results to ensure that tool responses are consistent, and provides verification that the tool is operating correctly.

To evaluate the density tool response to heterogeneity in the vertical dimension, or the so-called thin-bed resolution response, a log of measurements along a large, abrupt change in the formation density is used to determine the vertical resolution in the density response. Computer modeling then is used to evaluate the tool's response to heterogeneity in the radial direction, such as that caused by formation damage or variations in the fluid density in the invaded zone between the borehole wall and the deeper formation. The two response functions are folded into an algorithm employed during analysis of the logging data.

Neutron Porosity Calibrations. Neutron porosity tools are sensitive to a wide range of environmental effects including borehole size, mudcake thickness, borehole salinity, mud density, borehole temperature and pressure, as well as lithology. As such, a complex primary tool calibration is performed.

Primary tool calibrations are completed with a minimum of three different lithologies (e.g., limestone, dolomite, and sandstone) in at least six borehole sizes ranging in diameter from 6 to 24 in. Calibration measurements are completed in formations of zero, low, and medium porosity, and a 100% water tank for each borehole size and lithology with different mudcake thicknesses and weights. The number of configurations needed to define the basic response calibration in these environmental exceeds 250 tank measurements.

Salinity also has a large effect on the thermal neutron porosity response, both in the borehole and formation. These measurements add another factor of approximately eight, bringing the total number of primary calibration measurements to approximately 2,000. Even with this large number of measurements, not all possible configurations were tested. Temperature effects are tested with tank measurements and computer modeling (at high temperatures from 100 to 175°C where the water in laboratory tanks would boil).

Environmental corrections are essential to accurate porosity measurements. In the presence of clay materials or hydrous alteration minerals, a correction is required to account for the presence of bound water. Because the hydrogen is present not only as free water but also as bound water in clay minerals, the porosity response, often combined with the density log, can be used to detect shaley intervals or other minerals (e.g., gypsum) that have a high hydrogen index because of its water of crystallization. Conversely, the neutron curve can be used to identify anhydrite and salt layers, which both are characterized by low neutron readings and by high- and low-bulk density readings, respectively.

Hole size affects neutron log responses; in particular, the formation signal for epithermal count rates tends to be masked by the borehole signal with increasing hole size. In liquid-filled holes, the influence of the borehole fluid depends on its salinity. Chlorine is a strong neutron absorber and has a large impact on thermal neutrons. Additionally, density has an impact on porosity measurements, and the addition of weighting additives (e.g., barite) will yield lower porosity readings. Most porosity tools are used in open-hole applications in the oil and gas industry; however, the DOE and the Office of Assistant Manager for Development and Production sometimes log through the drilling pipe and the bottom hole assembly. Since iron is a strong neutron absorber, the effect is that of an increased porosity reading, depending on the thickness of the pipe. As such, additional environmental corrections must be implemented to account for the effects of the drilling pipe.

Accelerator porosity tool calibration measurements include studying the effects of tool standoff as well as the other effects mentioned above. A simulated shale formation was developed using aluminum oxide as the porous formation media to determine the effects of high-density shale on this tool's response.

As with density tools, an evaluation of neutron tool response to heterogeneity in the vertical dimension (thin-bed resolution response) was done with a series of measurements using abrupt, large changes in the formation porosity. Computer modeling also has been used to evaluate the tool's response to heterogeneity in the vertical and radial directions.

Shop calibrations are performed with precision-built water tanks that provide single-point-calibrationsfor the porosity response for the CNT-G (small tank) and APT (large tank). Well-site checks for the CNT-G tools are done with a small (61 cm [2 ft] long) precision-built block that contains an organic material and a weak neutron source to produce a calibration count rate in the tool's detectors that simulate logging environments. Well-site checks for the APT consist of measuring passive count rates from tiny alpha-n sources built into the detectors and by switching on the accelerator once the tool is safely lowered into the well. A repeat pass of one section of the well is used to compare the logging results to ensure that the tool responses are consistent and helps verify that the tool is operating correctly.

Passive Gamma-Ray Calibrations. Primary tool calibrations for natural gamma-ray tools usually consist of running the tools in large, man-made formation pits with a carefully mixed amount of radioactive material in base media, usually sandstone or cement. Unfortunately, many of the standard formation test facilities, such as the American Petroleum Institute (API) standards at the University of Houston, have been found to be inaccurate. This has forced the logging service companies to develop their own natural radioactivity calibration formation test pits. Typically, one pit is enriched in each naturally radioactive element (e.g., potassium, uranium, and thorium) and a fourth one that has a mix of all three elements to simulate typical radioactive shale. The large well-logging companies (e.g., Schlumberger) maintain their own set of natural radioactivity calibration standards. Calibration facilities for gamma-ray and fission-neutron well logging measurements also exist at the DOE Grand

Junction Project Office in Grand Junction, Colorado, and at the DOE Hanford Site near Richland, Washington. These facilities consist of buried concrete models with enhanced, known amounts of potassium, uranium, and thorium, and are used to calibrate logging tools to the gamma rays emitted by these elements. These facilities will be described hrther in Section 3.

The gamma-ray intensity from a uniformly distributed source of constant mass concentration is independent of the formation density, even though the attenuation is a direct hnction of the formation density. However, passive spectral measurements suffer to some degree from the borehole environment. Gamma rays emitted from the formation have to pass through different amounts of absorber materials to reach the detector, based on the quantity of mud in the borehole and varying hole diameters in fluid-filled boreholes. Additional complications arise from the natural radioactivity contained in common, dense-mud additives (e.g., barite and KCl). Therefore, environmental correction calibrations are required for borehole size and fluid, casing effects, and mud. These environmental corrections are provided by Monte Carlo models that are bench marked using measurements in the calibration standards.

Fabric sleeves containing a mixture of naturally radioactive clays, which can be wrapped around the NGT tool at the detector location, serve as both shop and well-site calibrations. A standard practice is where a repeat pass of one section of the well is used to compare the logging results to verify that the tool responses are consistent and that the tool is operating correctly.

Pulsed-Neutron Thermal Decay Time Calibrations. Primary tool calibrations for pulsed-neutron thermal decay-time logging tools consist of evaluating the tool response over the complete range of possible water salinities in the borehole and formation, and over a wide range of formation porosity, borehole size, and differing lithologies. Additionally, temperature and pressure effects can influence the decay time measurement; as such, appropriate corrections are typically quantified with benchmarked results derived from the Monte Carlo model. Thousands of primary tool calibrations are made in the test tank formations, and are used to build a library of tool responses that form a base network of parameters for a regression interpolation.

As with the neutron porosity tools, a precision-built water tank is used for shop calibration of the thermal neutron decay time measurements. Examining detector count rates after these tools are first lowered into the well satisfies the well-site calibration. A repeat pass of one section of the well ensures that the tool responses are reproducible and helps verify that the tool is operating correctly.

Appendix B

Alternative Measurement Systems

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Alternative Measurement Systems

Above-Ground Measurement Systems

Long-Range Alpha Detector. The (long-range alpha detector) LRAD technique relies on ionization of ambient air molecules by alpha particles. These ions are transported to an ion collector where the small current produced by this ion flow is measured with a sensitive electrometer. This current is proportional to the number of ions and hence to the strength of the alpha contamination averaged over the entire surface from which the air is collected. Thus, LRAD-based monitors do not depend on direct detection of alpha particles themselves. The instrument is actually sensitive to all forms of ionizing radiation, but is best suited to detecting alpha particles because of their short range and dense ionization pattern. Beta particles and gamma rays have a much longer range and deposit most of their energy in the chamber walls. Neutrons interact weakly and do not produce a large signal in the LRAD. In most cases, the contribution from other types of radiation is negligible compared to the alpha contribution.

The LRAD-based monitoring systems are limited by the distance that the ions can travel through air (several meters) or soil (less than a few centimeters). To enhance measurements in soil at depth, it was once proposed to force high-pressure air through soil to carry ions to the detector; however, investigation of this concept was very limited and a field-scale system was never developed.

An LRAD soil-surface monitor was developed and demonstrated. It consists of a large (1 m^2 x 20 cm thick [1.2 yd^2 x 7.9 in.]), flat, box mounted on a tractor where the open end is placed on top of the soil. The tractor moves the detector between monitoring positions. About 15 minutes are required for signals to stabilize after the detector is moved to a new position. Once signals are stable, currents are averaged for 5 minutes. Radiation from the soil ionizes the air in the box. The amount of ionized air, which is proportional to the amount of radiation, is measured.

Because only alpha activity is measured and individual alpha-particle energies are not determined, the LRAD does not identify specific radionuclides. For this reason, the LRAD cannot distinguish between alpha activity from naturally occurring uranium, thorium, or man-made plutonium. Nor can it directly distinguish between alpha contamination and radon gas that emanates from the soil and mixes with air within the LRAD chamber. The LRAD surface monitor indirectly corrects for radon by incorporating a second compensating ion chamber that is exposed to air without direct exposure to the soil. Once air in the two ion chambers equilibrates, the difference between the signals is a measure of the soil alpha-contamination level. However, the seal between the LRAD chamber and soil is not always adequate, especially on ground that is uneven, which prevents signal stabilization.

A major source of uncertainty for LRAD measurements is soil conditions. The moisture content of soil can change dramatically under sun or precipitation conditions. This strongly influences LRAD performance because of related changes in soil outgassing, radon levels, and alpha penetration length. Also, the LRAD cannot be used in areas with high radioactivity levels because the rapid recombination of ion pairs created by the alpha radiation gives a false indication of the level of alpha activity present.

Several limitations of the LRAD technology are relative to application at the SDA. Most significant is the very limited depth of detection in soil, which is only a few centimeters. The depth of the overburden at the SDA will prevent detection of any alpha activity in the buried waste and removal of the overburden to expose the waste for purposes of performing a surface survey is not a viable option. Another limitation is the inability of the LRAD to distinguish between alpha activity from uranium and plutonium. Much of the buried waste at the SDA contains significant quantities of uranium, which is not considered a transuranic element. Thus, there would be no means of distinguishing a uranium hot spot from a TRU hot spot. In addition, radon is in the gas emanating from the SDA; therefore, a high probability of false-positive readings exists because of radon.

Long-Range Neutron Detection. Transuranic isotopes generate neutron emissions, either from spontaneous fission or (α,n) reactions on light isotopes. Consequently, the neutrons can be usehl for quantifying TRU when copious fission or activation products mask their gamma emissions. Both thermal and nonthermal neutrons can travel long distances in air, making remote detection possible. The incident neutron spectrum depends both on the original (source) spectrum and on the moderation that occurs through neutron interaction with air and soil. Increased scattering of the neutrons tends to increase the thermal portion of the spectrum while reducing the directional information.

A neutron detector designed for detecting neutron sources at distances of 50 to 100 m (55 to 109 yd) in air was constructed and tested by the Pacific Northwest National Laboratory. This detector has a large surface area (1 x 1 m [3.2 x 3.2 ft]) to enhance detection efficiency, and it contains a collimator and shielding to achieve direction sensitivity and reduce background. Test results show that moderated fission-neutron sources of strength (about 3×10^5 n/seconds) can be detected at a distance out to 70 m (77 yd) in a counting time of 1,000 seconds. The best angular resolution of the detector is obtained at distances of 30 m (33 yd) or less. The environment through which the neutrons pass to reach the detector does moderate the fast neutrons and significantly affects the energy spectrum incident on the detector. As the separation distance between the source and detector increases, the contribution of scattered neutrons to the measured signal increases. This results in a decrease in the ability to detect the direction to a distant source. The intended applications of the long-range neutron detector include detecting remote neutron sources (including sources in moving vehicles), monitoring source storage vaults, measuring the TRU content of waste in the presence of high gamma-ray background.

A major limitation of the long-range neutron detector relative to use in the SDA as a surface monitor to determine the distribution of TRU waste and identify TRU hot spots is the limited range of neutrons in soil, especially when the soil is moist. The transit of neutrons through dry soil can be a much as 1 m (3.2 ft), but the transit through water-saturated soil could be less than a few centimeters. As such, the depth of the overburden [approximately 1.5 m (5 ft)], exceeds the range of neutrons in soil. Additionally, neutron-absorbing elements in the buried waste (e.g., chlorine) also will serve to attenuate the neutrons emitted in the waste. Although the neutron-absorbing material in the waste cannot be easily removed, the overburden could be excavated to a reasonable depth (i.e., 30 cm [1 ft]). Removal of the overburden to within 30 cm (1 ft) of the waste for purposes of surface monitoring would be of limited value, as detection of neutrons from TRU in the waste would be limited to the top 30 to 60 cm (1 to 2 ft) of waste. Significant quantities of TRU below this depth would not be detected.

Prompt Gamma Neutron Activation Analysis. The method of prompt gamma neutron activation analysis (PGNAA) is potentially a powerful method for noninvasive and rapid determination of vertical contaminant profiles in soil. In this method, fast neutrons are directed into the soil from a plane-parallel beam or from an isotropic point source on the surface. A collimated radiation detector above the soil then measures intensities of the characteristic gamma rays released by elements in the soil that capture the neutrons. Analytical procedures for deconvoluting or inverting the measured gamma-ray spectra then are applied to estimate contaminant concentration profiles.

The experimental N-SCAN system, based on PGNAA, was developed by Westinghouse Electric Corporation for the U.S. Department of Energy to provide near real-time subsurface characterization of radioactive contamination in soil and concrete. The system combined a specially modified electronic neutron-generator tube, high-efficiency gamma detection, and computer-directed microsecond control capability of the source and detection process. Incident neutrons initiate nuclear reactions in atoms of the sample matrix and contaminants. These nuclei emit energetic "prompt" gamma rays in 10 to 14 seconds, which are the signatures of the original target atom-matrix or contaminant.

In the survey system, a correlated, computer-controlled sequence of neutron emission and gamma detection was employed to produce data with high signal-to-background ratios and for trace contaminants in massive matrices (e.g., soil or concrete). This was coupled with measurement capability using algorithms produced from benchmarked radiation transport modeling to attempt near real-time, in situ trace-level-contamination characterization, including depth profiles. Further development of the N-SCAN ceased in 1995 after it was determined that the system could not provide satisfactory detection limits necessary for SDA remediation.

As with the LRAD and the long-range neutron detector, the depth of the overburden at the SDA is a severely limiting factor for a PGNAA surface monitor. The depth of the overburden soils greatly exceeds the penetration limit of neutron and gamma rays. However, development has proceeded on PGNAA technologies for soil characterization focused on borehole applications that show promise for effective application at the SDA.

New Probe Development for Transuranic and Mixed Low-Level Waste Analysis

In Calendar Year 2000, the Idaho National Engineering and Environmental laboratory initiated development of two advanced nuclear tools to perform in situ characterization of buried waste at the SDA to eliminate the risk of exposing personnel to contaminants or generating new waste. The tools discussed below will be operated within the Type A probes.

Prompt-Fission Neutron Tool. The first tool under development is an enhanced prompt fission neutron (PFN) device that uses a special zero-background pulsed-neutron generator to induce thermal-neutron fission in TRU waste elements (e.g., Pu-239). The fission of Pu-239 results in production of epithermal neutrons that the probe can detect in the time interval between neutron pulses from the generator. The measurement of epithermal neutrons is an extremely sensitive and specific signature of fissile material in materials surrounding the tool. The concentration of fissile material in the subsurface soil and buried waste is determined from the intensity of detected epithermal neutrons. A dual-spaced epithermal neutron detector array will measure moisture level. Thermal neutron and sodium iodide gamma-ray detectors will determine the neutron absorption cross section in the subsurface materials. Moisture content and neutron absorption cross-section measurements are needed to make corrections to observed fission neutron count rates caused by variations in environmental conditions. In a passive mode, the PFN detector system can measure neutrons produced from spontaneous fission to identify sources of fissile material detected in the buried waste.

Pulsed Neutron-Gamma Tool. The second tool under development is a pulsed neutron-induced gamma-ray spectrometer with high-resolution spectral capability. This device can measure elemental concentrations from neutron-induced gamma-ray emissions and from the decay of radioactive isotopes such as Pu-239, Am-241, and Cs-137. Carbon concentrations in the waste can be determined by measuring the inelastic-neutron scattering. Neutron capture and activation reactions are used to measure a suite of elemental concentrations, including TRU and chlorine. The pulsed neutron-induced gamma-ray spectrometer passive and capture measurements will identify and quantify TRU elements in the buried waste. Sensitivity of the gamma-ray spectroscopy, both passive and active, is limited because of shielding by soil, waste materials, and the steel casing.

These tools were first field tested at the SDA in September 2001. Currently, calibration and laboratory experiments are being conducted on the tools to better understand environmental effects on measurements at the SDA and to establish detection limits.